

TITLE: Development of Superior Sorbents for Separation of CO₂ from Flue Gas at a Wide Temperature Range during Coal Combustion: IC Phase II

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1. ABSTRACT

Program Introduction: Rationale and Objectives

CO₂ is considered as the major greenhouse gas contributing to global warming. The use of metal exchanged low silicon (LSX) faujasites is widely accepted in industry but it is limited by the fact that it can be used for mixtures containing only low concentrations of CO₂. Unfortunately, those sorbents are effective to remove CO₂ from gas mixtures which only contain gases that are less polar than carbon dioxide. Existing sorbents cannot operate at elevated temperatures due to rapid loss of structural stability and regenerability. Moreover, the existing systems are not versatile to be utilized for other processes (i.e. fuel cell applications, purification of H₂ from WGS, membranes, etc.) and they are also very sensitive to poisons and impurities.

Hence, a new generation of sorbents for removal of CO₂ at a wide range of temperatures (30 to 650 °C) is needed. The high temperature can be utilized for the CO₂ removal from hot gas streams (flue gas, gas generated from coal gasification, fuel cell applications, etc.). Equally important requirements are the high temperature stability of the sorbents as well as their ability to be regenerated reversibly and restore their performance to the initial levels. Sorbents with high selectivity for CO₂ with respect to other gases of multicomponent mixtures (including gases which are more polar than CO₂) is *a must for* an effective separation. Successful sorbents should have low affinity for water, O₂, and N₂ and they should be tolerant to poisons and should demonstrate ease for adaptability into existing units.

The objectives of this project are:

- 1) Integrate the synthesis efforts aiming at developing basic and superbasic supports and sorbents. Enhance the knowledge of functionalizing the surface for obtaining the desirable performance.

- 2) Characterize the synthesized sorbents with numerous state-of-the-art characterization techniques to understand their surface properties and functionality.
- 3) Direct the synthesis and characterizations toward highly durable systems that will ensure stability for regeneration over many hundred cycles at elevated temperatures and severe operating environments.
- 4) Use the obtained information as feed-back to design the most effective sorbents. Test these sorbents with gas mixtures containing CO₂, H₂O, air, SO₂, SO₃, H₂, and CO at selected compositions simulating industrial operation.

Accomplishments Achieved during the Current Period of Performance

We have developed high temperature Cs/CaO sorbents **for CO₂ separation with zero affinity for N₂, O₂ and NO**. The carbonation rate of these sorbents was improved reasonably by a full oxidation of the cesium support. The presence of water vapor significantly promotes the CO₂ sorption capacity of the sorbent.

Moreover, we found that calcium oxide sorbents synthesized from calcium acetate monohydrate exhibited high carbonation rates (about 98% of the theoretical value) under cyclic operation.

We have initiated the synthesis of aerosol made sorbents which possess high surface area and high carbonation at 700°C. One of the main characteristics of the aerosol made versus the sol-gel made sorbents is their durability. This property is essential for the synthesis of superior industrial sorbents for extensive operation.

Plans for the coming year

- Correlate sorption performance of the sorbents under investigation with surface characteristics for obtaining a better fundamental understanding.
- Synthesize aerosol CaO-based sorbents with specific surface area. Use selected dopant to reinforce their structure.
- Characterization of the sorbents with state-of-the-art analytical techniques.
- Continue sorption experiments with the new family of sorbents. Use realistic gas mixtures (N₂, O₂, H₂O, NO and SO₂) simulating flue gas composition.

2. LIST OF PUBLISHED JOURNAL ARTICLES, COMPLETED PRESENTATIONS, AND STUDENTS RECEIVING SUPPORT FROM THE GRANT

Journal Articles

- 1) E.P. Reddy, and P.G. Smirniotis. "High-temperature sorbents for CO₂ made of alkali metals doped on CaO supports", *Journal of Physical Chemistry B*, (2004), 108 (23), 7794-7800.

Journal Articles (in press)

- 1) A. Roesch, E.P. Reddy, and P.G. Smirniotis. "Parametric Study of Cs/CaO Sorbents with respect to Simulated Flue Gas at High Temperatures", *Industrial Engineering Chemistry Research*, in press, (2005).

Conference Presentations

Students Receiving Support from the Grant.

Graduate Students:

- 1) Mr. Hong Lu, graduate student (Ph.D.) in Chemical Engineering